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Catalysis Today 98 (2004) 265–271
CATALYSIS TODAY
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Effect of high pressure operation on overall phase holdups in ebullated-bed reactors

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Available online 15 September 2004

Abstract

The effect of pressure on basic hydrodynamics of ebullated beds at elevated pressures of up to 15 Mpa was studied by measurement of bed profiles by gamma-ray densitometry. For 1.7 mm glass beads the bed expansion as well as gas holdup increased with pressure, while liquid and solids holdup decreased. Given the solids holdup is available, it was found that the generalized bubble wake model could be applied to compute the liquid and gas holdups of ebullated beds at high pressures. The mean absolute error of the calculated liquid holdups for the present work and for literature data was of the order of 5%.

Keywords: Ebullated beds; Phase holdups; High pressure

1. Introduction

Ebullated-bed reactors are used in several hydroprocessing technologies for upgrading residual materials. These processes operate at high severity and can treat heavy feedstocks of different characteristics to produce maximum distillates and minimum fuel oils.

In an ebullating bed reactor, the heavy oil feed stock, hydrogen and the catalyst particles are brought together in constant contact and motion. The catalyst particles are suspended in the heavy oil and are fluidized by a constant flow of liquid and gas from the bottom of the reactor. Reactor performance is affected by the degree of contact achieved between phases and hence by the hydrodynamics of the bed. Hydrodynamic properties of these reactors at operating conditions are required for reliable design and operation. Among the most important macroscopic hydrodynamic parameters used for characterizing an ebullating bed are the three phase holdups, ε_g, ε_l, and ε_s, each of which respectively represents the bed volume fraction occupied by the gas, liquid and solids phases. Solids holdup can be obtained from bed expansion measurements, which appears to be is a parameter that is often measured in ebullating bed reactors for control purposes.

Extensive experimental work has been devoted to the study of fluidization properties at atmospheric pressure and room temperature, however, hydrocracking takes place at temperatures of 420–450 °C and pressures in the range 100–200 bar, depending on feedstock. Information describing these three-phase systems at high severity conditions is comparatively scarce due to measurement difficulties. Only few works in the open literature have explored the nature of pressure effects on the basic hydrodynamic behavior. Jiang et al. [1] combined visualization in two-dimensional operation and holdup measurements in three-dimensional and reported that with increasing pressure from atmospheric up to 1 MPa bubble size decreases, bubble size distribution narrows, and gas holdup increases. Luo et al. [2] studied the effect of pressure over the range of 0.1–15.6 MPa and found that the bubble-size reduction produced by an increase in pressure leads to an increase in the transition gas velocity

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0920-5861/$ – see front matter
doi:10.1016/j.cattod.2004.07.039

from the dispersed bubble regime to the coalesce bubble regime, an increase in the in the gas holdup, and a decrease in the liquid and solids holups. It was also found that the pressure effect was insignificant above 6 MPa. Jiang et al. [3] investigated the effect of high temperature and pressure over bed expansion and contraction and reported that the effect can be characterized by the changes in bubble behavior and the liquid properties. They found that the generalized wake model of Bhatia and Epstein [4] could account for bed expansion or contraction phenomena by taking into account bubble size distribution effects.

For prediction purposes, although there are several empirical published correlations for calculating phase holdups, they have been derived basically from low pressure data and hence they seem to be inadequate for ebullating bed reactors operated at hydroprocessing conditions. Bhatia and Epstein [4] developed the generalized wake model to analyze the bed porosity with the volume ratio of wake to bubble. Although this model has been verified well with data obtained mostly under ambient conditions, results such as those of Jiang et al. [3] suggest that it could also be useful for describing systems operated at high temperature and pressure conditions.

In the present work the bed expansion characteristics of an ebullating bed operated at high pressures is utilized for predicting the corresponding gas and liquid holdups. Experimental data on gas, liquid, and solids phase holdups are presented in a broad pressure range for different particle size and gas and liquid superficial velocities. The analysis of the data is based on the generalized wake model, which considers the influence of gas bubble wakes on bed expansion and liquid holdup behavior.

2. Experimental

The reaction section of a hydrotreating pilot plant was utilized in the determination of experimental hydrodynamic data at high pressure conditions (see Fig. 1). The reactor was an ebullating bed 434 cm high and 2.94 cm in diameter. It has been design to operate at severe conditions of temperature and pressure and so pressures of up to 15 MPa have been considered. The bed consisted of 1.71 mm diameter glass beads, 2509 kg/m^3 density, supported by a wire mesh placed at the bottom of the reactor, fluidized by co-current diesel fuel ($\mu_l = 45 \times 10^{-4}$ kg/ms, $\sigma = 30 \times 10^{-3}$ N/m, and $\rho_l = 836$ kg/m^3 at 20 °C and 0.1 MPa) and nitrogen flows. Bed density profiles were obtained with a commercial gamma-ray densitometry SGD Density System unit from TN Technologies. The technique is totally non-invasive and uses energy attenuation to measure density. As the overall density of the bed changes, so does the energy that penetrates the bed. The system basically consists of a nuclear density gauge mounted in a movable assembly mechanism, which was used to transport the gauge axially along the column. The gauge is comprised of a Cesium-137 source contained in a lead-filled, steel-encased housing mounted on one side of the bed, and a scintillation detector which is mounted on the opposite side of the column. For calculation of the bed density at a given height, the system has to be previously calibrated with a fluid of known density, and then it utilizes the attenuation coefficient of the process material components to compute the response to process density changes. From the bed density profiles the average bed densities, ρ_B, were determined. The expanded bed heights, H_B, were obtained from the position where there was an abrupt change in the column axial density profile, which corresponded to the density change at the bed-freeboard interface.

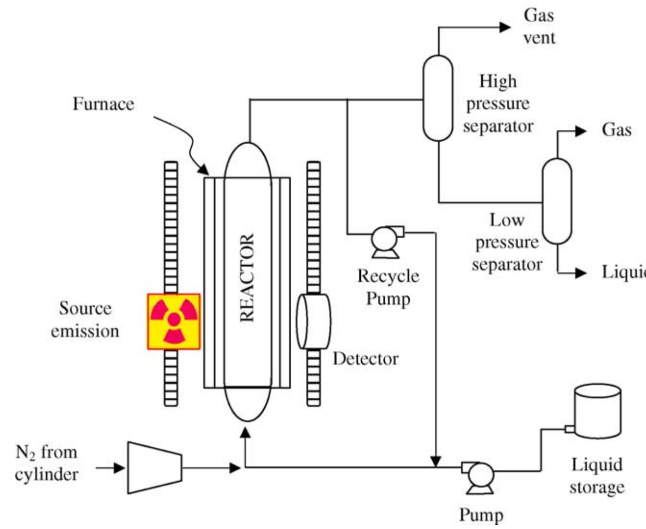

Fig. 1. Schematic diagram of the experimental system.

The overall solids holdup is related to the expanded bed height by

$$\varepsilon_s = \frac{4M_p}{\pi D_t^2 \rho_s H_B} \tag{1}$$

The liquid and gas holdups are then determined from solving the following equations

$$\rho_B = \rho_s \varepsilon_s + \rho_l \varepsilon_l + \rho_g \varepsilon_g \tag{2}$$

$$\varepsilon_g + \varepsilon_l + \varepsilon_s = 1 \tag{3}$$

3. Results and discussion

3.1. Effect of pressure on phase holdups

Pressure is found to have a significant effect on the hydrodynamics of ebullating bed reactors. The gas holdup is affected through the variations in the bubble characteristics and the flow regimes. The average bubble size decreases and the bubble size distribution becomes narrower with an increase in pressure [2]. The bubble-size reduction leads to an increase in the transition gas velocity from the dispersed bubble regime to the coalesced bubble regime. For systems in the dispersed bubble regime where both gas holdup and bubble rise velocity are uniformly distributed in the bed, the gas holdup and bubble rise velocity are related by [5]

$$\varepsilon_g = \frac{U_g}{U_b} \quad (4)$$

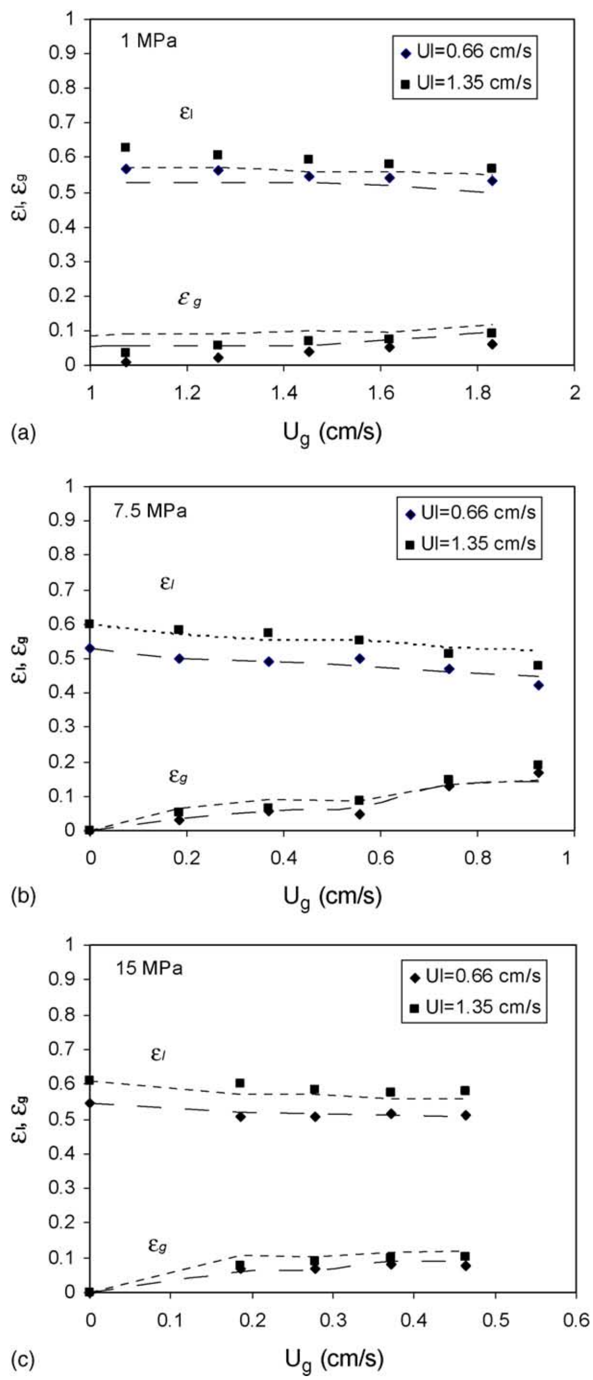

Fig. 2. Effect of gas velocity for various liquid velocities and (a) P = 1.0 MPa; (b) P = 7.5 MPa; (c) P = 15.0 MPa.

Since bubble rise velocity, U_b, decreases with bubble size, it is clear from Eq. (4) that higher gas holdups can be attained in high pressure beds where small bubbles prevail than at systems operated at low pressure, were relatively larger bubbles are frequent.

Fig. 2 shows the experimental results of gas and liquid holdups at different pressures. It was found that at 1.0 MPa, the lower pressure considered, the bed gas holdup was always lower than when it was operated at higher pressures. The liquid holdup, on the contrary, was found to decrease with an increase in pressure. According to visual observations of high pressure three-phase systems, pressure suppresses bubble coalescence and enhances bubble breakage by particles, furthermore, it has also been observed that the distributor tends to generate smaller bubbles under extreme pressure conditions [2]. All these factors promote the existence of small and relatively slow bubbles that enhance gas holdup. The main effect of pressure on gas holdup, however, appears to occur at lower pressures since, as reported by Luo et al. [2], at higher pressures its effect seems to level off. They found a significant increase in gas holdup up to approximately 6 MPa, but that at higher pressures the increase was substantially reduced. The gas holdup results presented in Fig. 3 seem to be in accordance with such observations, as the gas holdups for 7.5 and 10 MPa appear to be of similar magnitude compared with those for 1 MPa.

Bed expansion or solids holdup has been widely studied under ambient conditions. For such systems it has been well established that bed expansion increases with an increase in the liquid velocity. Furthermore, upon introduction of gas into a liquid–soild bed the bed can either expand or contract, depending on the particle properties and liquid velocity. Similar behavior has also been reported for ebullating beds operated at high pressure [3]. The effect of pressure on the extent of bed contraction or expansion for 1.71 mm glass beads as a function of gas velocity is presented in Fig. 4. As it can be seen, for both liquid velocities considered the bed expands under all pressure conditions. Similar results were reported by Luo et al. [2] for 2.1 and 3 mm glass beads at low liquid velocities, however, bed contraction was observed for systems with these particles at a relatively high liquid velocity of 2.6 cm/s. It is also apparent that for given gas and liquid velocities that the bed expands with an increase in pressure.

In ebullating beds particles are suspended by both the liquid flow and the flow induced by bubble motion. The effects of pressure on bed expansion or contraction can be attributed to their effects on the physical properties of the liquid and on bubble behavior. The liquid properties directly affected are mainly viscosity and density. Bubbles under high pressure conditions tend to be smaller in size than those at low pressures, these both, reduce the amount of liquid entrained by the bubble wakes and increases the gas holdup, both of which work towards an increase in the bed expansion.

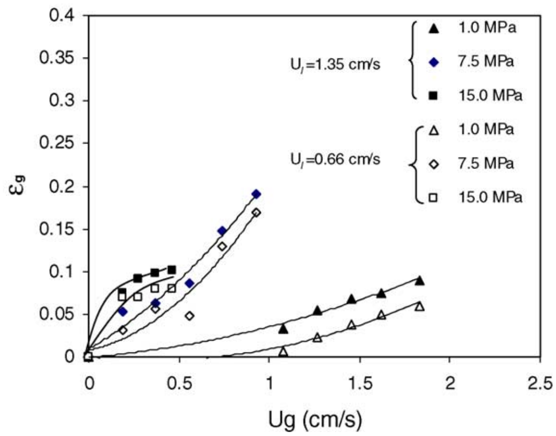

Fig. 3. Effect of gas velocity on gas holdup for various values of pressure and liquid velocity.

3.2. Mathematical model

In an ebullated bed extreme pressure and temperature operating conditions can have an effect on bubble properties which, in turn, can impact bed macroscopic hydrodynamics as bed expansion and phase holdups [1–3]. A key factor that has been recognized in explaining various phenomena occurring in three-phase fluidized beds consists of the bubble wake. Wake models like the generalized wake model developed by Bhatia and Epstein [4] have proved to be widely accepted for design purposes [5] and so this model will be utilized in the present work. In the generalized wake model the bed is subdivided into a liquid–solid fluidized region ($\varepsilon_{l-s} = 1 - \varepsilon_g - \varepsilon_w$), a gas bubble region ($\varepsilon_g$) and a wake region ($\varepsilon_w$). Among the main assumptions of the model are: (i) the solids content in the wake can be an arbitrary value different from that in the liquid–solid fluidized region; (ii) the wake rises at the same velocity as that of the bubble; (iii) the Richardson-Zaki correlation between liquid velocity and solids holdup applies in the liquid–solid fluidized region [6].

In the generalized wake model, the key parameters are the ratio of the bubble wake volume to the bubble volume, k, and the ratio of the solids holdup in the wake to the solids holdup in the liquid–solid region, x, are written as

$$k = \frac{\varepsilon_w}{\varepsilon_g} \tag{5}$$

and

$$x = \frac{\varepsilon_{sw}}{\varepsilon_{sf}} \tag{6}$$

respectively. The liquid and solids linear velocities in the liquid–solid fluidized region, V_{lf} and V_{sf}, respectively, and their relationship in terms of a Richardson-Zaki type of expression can be written as [5]

$$V_{lf} = \frac{U_{lf}}{\varepsilon_{lf}} = \frac{U_l - U_g k[1 - x(1 - \varepsilon_{lf})]}{\varepsilon_{lf}[1 - \varepsilon_g(1 + k)]} \tag{7}$$

$$V_{sf} = -\frac{U_g k x}{1 - \varepsilon_g(1 + k)} \tag{8}$$

and

$$V_{lf} - V_{sf} = U_i(\varepsilon_{lf})^{n-1} \tag{9}$$

where U_i is the extrapolated superficial liquid velocity as $\varepsilon_l \to 1$ in the liquid–solid region and n is the Richardson-Zaki index. The velocity U_i can be estimated with the following equation [7]

$$U_i^{1.4} = 0.072\frac{d_p^{1.6}(\rho_s - \rho_l)g}{\rho_l^{0.4}\mu_l^{0.6}} \tag{10}$$

There are several correlations in the literature for the estimation of the ratio of the bubble wake volume to the bubble volume, k, and the ratio of the solids holdup in the wake to the solids holdup in the liquid–solid region, x [4,8–10]. Most correlations available are purely empirical and precautions should be taken whenever used. From a comparison between several correlations those of Bhatia and Epstein [4] and Baker et al. [10] have been recommended [5]. In their model Bhatia and Epstein [4] allowed the solids

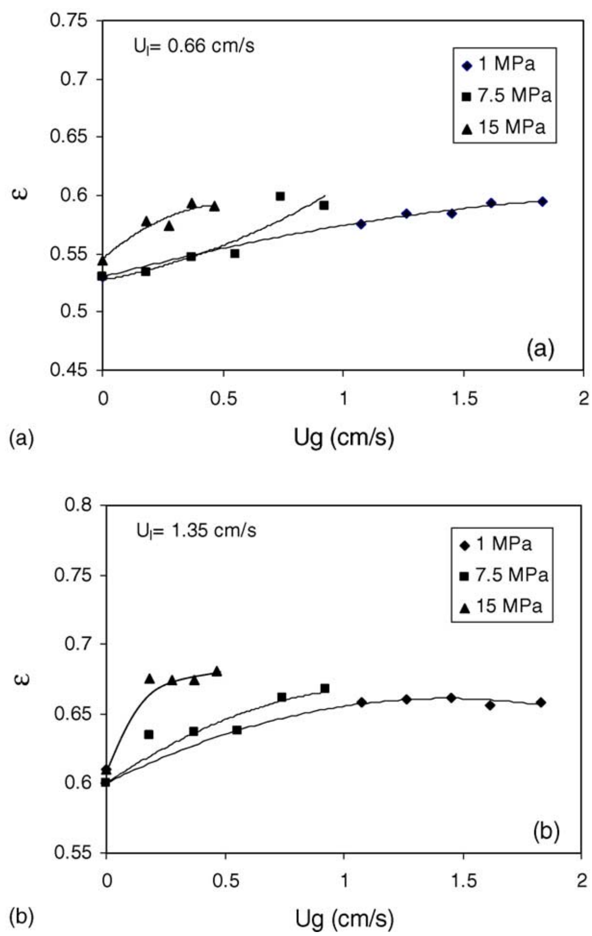

Fig. 4. Effect of gas velocity on bed porosity at various pressures and (a) U_l = 0.66 cm/s; (b) U_l = 1.35 cm/s.

concentration in the wakes to vary from zero to the value in the continuous phase. However, they concluded that provided the solids are wetted the assumption that the wakes are solid free is probably justified. They proposed the following expression for k (for $0 \leq x \leq 1$)

$$k = k_0'(1-\varepsilon)^3 \tag{11a}$$

and

$$k_0' = 0.61 + \frac{0.037}{\varepsilon_g + 0.013} \tag{11b}$$

Baker et al. [10] proposed an empirical correlation for k_0 and also for the bed porosity relationship in the liquid–solid fluidized bed in place of the Richardson-Zaki equation commonly employed in wake models. They proposed the following equation for k_0 (for $x = 0$)

$$k_0 = 0.01765\left(\frac{U_l}{U_g}\right)^{0.61}\sigma^{-0.654} \tag{12}$$

There is almost no evidence in the literature concerning the solids holdup in the wake of bubbles in fluidized beds operated at high pressure. Jiang et al. [3] analyzed the bed contraction criteria upon introduction of a gas flow in a liquid–soild fluidized bed at elevated pressures and found good agreement between simulations and experimental results when the wake was assumed to be solids free for large 2.1 mm particles. In the present work the solids holdup in the wake has also been assumed to be zero, i.e., $x = 0$. With this assumption the liquid holdup in the liquid–solid fluidized region can be related to the overall gas and liquid holdups by

$$\varepsilon_{lf} = \frac{\varepsilon_l}{1-\varepsilon_g(1-k)} \tag{13}$$

Eqs. (7)–(10) and (13), together with either Eqs. (11) or (12), can be solved, given one of the overall phase holdups is available, for the determination of the other two-phase holdups. In many practical cases the bed expansion level is a known process variable and with which the overall solids holdup, ε_s, can be calculated given the bed solids inventory and the particle density.

3.3. Simulation results of the model

The experimental system has been modeled with the generalized wake model, given the solids holdup, as described above. The gas and liquid holdups calculated for the data in Fig. 2 have presented as dotted lines in the same figure. For these calculations the ratio of the bubble wake volume to the bubble volume, k, the expression of Bhatia and Epstein [4] was utilized, however, it was found that for the operating conditions of the data in Fig. 2 very similar results were also obtained with the correlation of Baker et al. [10]. The Richardson-Zaki index, n, in Eq. (9) was obtained from the experimental data for the liquid–solid systems at the respective operating pressure. The liquid properties were estimated with HYSYS process simulator. The calculated liquid holdups in average were found to vary within 4% of the experimental values, and as can be seen from Fig. 2, relatively larger errors appear to exist at the lower pressure considered (0.1 MPa).

The model has also been utilized to calculate the liquid and gas holdups for the high pressure experimental conditions reported by Luo et al. [2], as shown in Figs. 5 and 6. The calculated values have been reported using both Bhatia and Epstein [4] and Baker et al. [10] correlations for calculating k. In general terms, for the operating conditions considered, the use of Bhatia and Epstein's equation (11) appears to produce calculated holdups closer to the experimental data. However, the use of this correlation at relatively high gas flow rates tends to produce larger predicting errors and can even yield negative values for V_{lf} in Eq. (7) unless the value of x is considered to be different from zero. Nevertheless, the mean absolute error of the calculated liquid holdups using the generalized wake model with Bhatia and Epstein correlation was of the order of 6% for the data in Figs. 5 and 6. The experimental liquid holdups of both the present work and that from Luo et al.

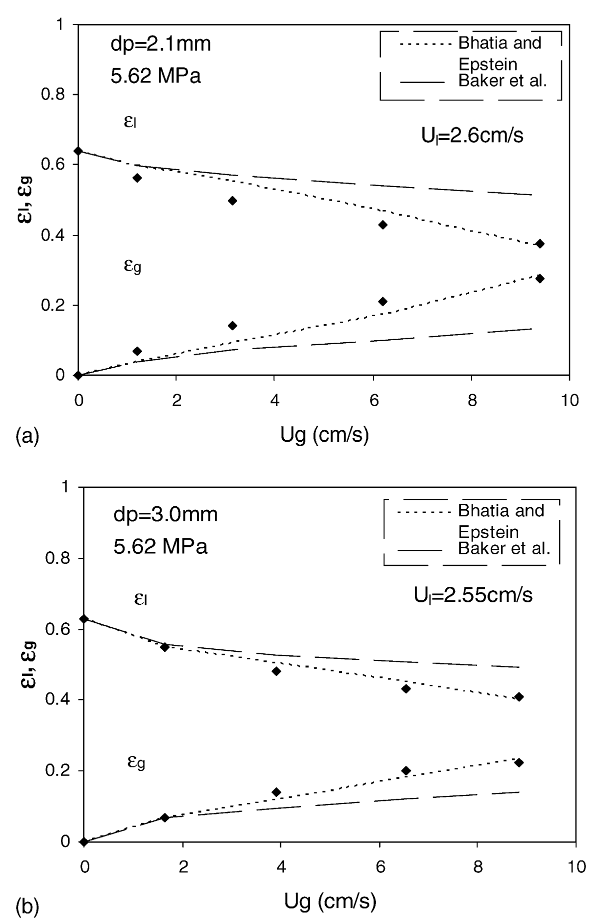

Fig. 5. Experimental holdups of Luo et al. [2] and calculated ones using k values from Eqs. (11) and (12) for (a) d_p = 2.1 mm; (b) d_p= 3.0 mm.

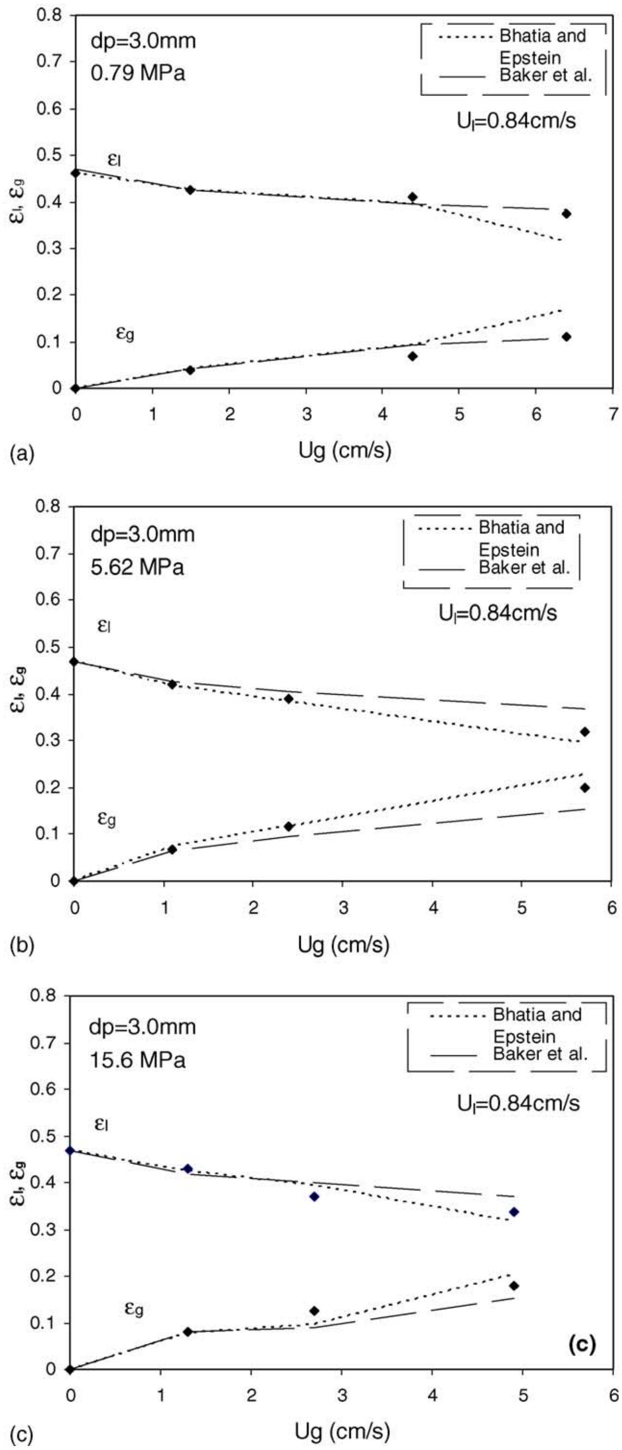

Fig. 6. Experimental holdups of Luo et al. [2] and calculated ones using k values from Eqs. (11) and (12) and (a) P = 0.79 MPa; (b) P = 5.62 MPa; (c) P = 15.6 MPa.

have been compared in Fig. 7 with the calculated values. It is clear from this figure that most of the predicted values are within a 10% difference of the experimental data and hence, the model appears to satisfactorily reproduce the experimental systems and to be a useful tool for estimating gas and liquid holdups at high pressure conditions when the

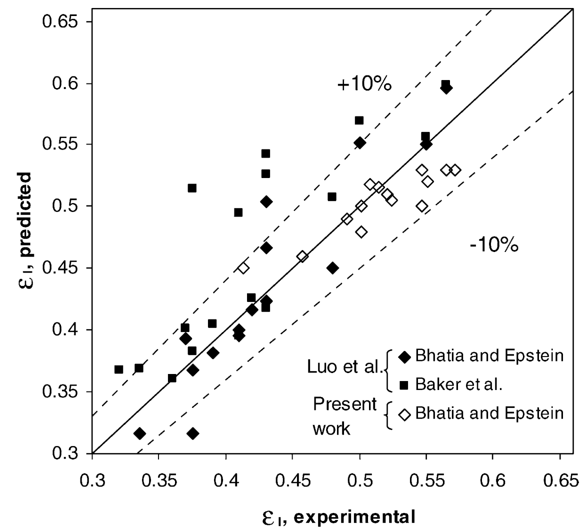

Fig. 7. Comparison of experimental data of Luo et al. [2] and of the present work with calculated values using k values from Bhatia and Epstein and Baker et al.

solids holdup is available or can be calculated from the bed height.

4. Conclusions

Pressure affects the hydrodynamics of ebullating bed reactors significantly by its effect on bubble behavior and liquid properties. For 1.7 mm glass beads fluidized with diesel fuel and nitrogen as the liquid and gas phases, respectively, an increase in pressure from 1 to 15 MPa was found to lead to an increase in the gas holdup and bed porosity and to a decrease in the liquid and solid holdups. The main effect of pressure however, appears to occur at lower pressures since at higher pressures its effect seems to level off. Similar results have been reported by other researchers for larger size particles [2]. The generalized bubble wake model of Bhatia and Epstein was utilized for describing the bed hydrodynamics of systems at high pressures. Its predictions compared favorably with both the experimental data of the present work and that of Luo et al. [2], and for which the mean absolute error of the calculated liquid holdups were 4 and 6%, respectively.

Acknowledgments

The authors thank Instituto Mexicano del Petróleo for the financial support.

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